# THE FIRST 10 YEARS OF SPACELAB MISSIONS: THE STRATOSPHERE AND GLOBAL CHANGE

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#### Abstract

The first ten years of Spacelab has seen it fly on a number of Space Shuttle missions carrying a myriad of science payloads including atmospheric remote sensing instruments. From low Earthorbit, these instruments have obtained measurements of the detailed composition of the atmosphere which have been crucial in improving our understanding of the stratosphere and its role in global change. This paper summarizes the results from one of these investigations, the Atmospheric Trace Molecule Spectroscopy (ATMOS) experiment, which has been involved in three Spacelab missions, and discusses some of the achievements in stratospheric measurements from this first decade of Spacelab flights.

#### Introduction

The 1970's saw increased scientific interest in studying the Earth's upper atmosphere and the coincidental announcement of opportunity for instruments for Spacelab missions on the Space Shuttle. While it had been proposed decades earlier that a kcy feature of the stratosphere was the presence of ozone, an absorber of a large amount of the solar ultraviolet radiation, the complexity of the photochemical processes which control ozone levels was just beginning to be recognized, with a dawning awareness also of the importance of those which mechanisms which lead to its erosion or depletion. These are commonly summarized in the catalytic cycles:

$$O_3 + X \rightarrow O_2 + XO$$
  
 $XO + O \rightarrow O_2 + X$ 

 $0_3 + 0 -> 202$ 

where X is NO, OH, or Cl

Toward the end of the decade, it was proposed that the growing amount of industrial chlorofluorocarbons (CFCs) released into the atmosphere, while effectively inert in the troposphere, would be photolyzed in the stratosphere releasing chlorine atoms, feeding directly into the catalytic destruction of ozone.

To understand the extent of the potential changes occurring as a consequence of these and similar anthropogenic activities required answers to three general questions:

what were the detailed processes which govern ozone levels?

what governs its geographic variability?

how will it change with time?

The challenges in answering these questions, in addition to the remoteness of the stratosphere, were the very low concentrations of the large number of gases thought to be important to understanding its photochemistry. If we could measure all of the ozone in the atmosphere as a pure layer of this gas at the surface, the ozone layer would only be around 3 mm thick, Spreading this layer out in the stratosphere, at heights above 15 km, leads to ozone concentrations of a few parts per million (ppmv, or parts per million by volume), while the gases which control its removal arc present at levels of less than a part per billion (ppby, or parts per billion by volume) down to parts per trillion (ppty, or parts per trillion by volume).

Detecting and measuring the cone cntrations of these different gases has required the development of a range of sensitive instrumentation to be used with many different carriers or platforms. However,

measurements from ground-based, aircraft or balloon-borne instruments arc limited by either their inability to look up through the denser troposphere to the stratosphere, or by their inability to provide global coverage at different geographic locations and conditions. The earlier free-flying satellite instruments were capable of providing global measurements, but were limited in capability as a result of weight, space, and resource limitations, These instruments were also plagued by problems due to their inability to sustain an accurate calibration standard over extended periods of on-orbit operation - a problem not limited to measurements of trends in stratospheric ozone, but also in determination of variations in the solar irradiance.

Mounting instruments on board the Space Shuttle overcomes many of these limitations. The key advantages of Spacelab are that it can support relatively large and sophisticated instruments, which can be flown, returned, refurbished, and recalibrated, then flown again in a series of such missions. In principle, these can be continued for many years beyond the normal lifetime of other satellite instruments, and on a time scale compatible with the longer term changes wc now believe to be occurring in the environment. Measurements by Spacelab instruments arc suited to three important areas: studies of stratospheric processes (chemical or physical); calibration of freeflying instruments with correlative measurements to validate trends within the Iong-term data sets; and repeating measurements over an extended period to establish an extended data set from which to estimate trends directly.

Onc instrument selected for the Spacelab missions was the Atmospheric Trace

Molecule Spectroscopy (ATMOS) Fourier transform spectrometer. While this is only onc of a number of atmospheric remote sensing instruments which have been flown on Space Shuttle, it typifies some of the kcy aspects of the role played by these instruments in studying the stratosphere. The primary objective of the ATMOS instrument

is to measure, simultaneously, the vertical profiles of more than thirty atmospheric constituents for use in studying stratospheric processes; these measurements are also useful in addressing the other two objectives of validation and trend determination.

#### **ATMOS**

In essence, the ATMOS experiment is based on a Fourier transform spectrometer capable of obtaining high resolution (0.01 cm<sup>-1</sup>) infrared solar absorption spectra, ATMOS gathers data in the solar occultation mode, twice an orbit when the sun sets or rises from behind the Earth. Most of the molecular species in the atmosphere absorb some portion of the solar infrared radiation, with a characteristic spectrum in the 2 to 16 lm wavelength range (600-4800 cm<sup>-1</sup>). Its weight of 250 kg, power consumption of 250 watts, high data rate and complex commanding requirements, make it an ideal candidate for the Spacelab environment. While technological advances in many areas could facilitate making a future ATMOS-like instrument smaller, lighter, and capable of being flown on a free-ftying satellite, an instrument this sophisticated and dataintensive presents other problems which are still more easily solved through its usc for brief periods on orbit at repeated intervals over an **extended** period of time. Further details about the current instrument are given elsewhere<sup>(1)</sup>.

The combination of the high spectral resolution coupled with the good signal-to-noise ratio achieved by using the sun as the light source provides ATMOS with the capability and the sensitivity with which to detect and measure many atmospheric trace gases. In order that these measurements be made with good vertical resolution (2 to 3 km), ATMOS records a spectrum every 2 seconds or about 100 total in a typical 4 minute observation period during sunset or sunrise. After some simple processing steps, sequences of transmission spectra of the Earth's atmosphere can be obtained which are the primary data product for analysis. A

sample sequence of spectra obtained by ATMOS on its first flight is shown in Figure 1. Data such as these, direct measurements of the infrared spectral response of the Earth's atmosphere, arc the strength of this particular experiment: they can be used to test and validate the real spectral response of instruments which do not have the same spectral discrimination without relying on theoretical data of limited quality; they are intrinsically or internally calibrated and therefore do not need elaborate calibration procedures, nor recalibrating for trends studies; they readily facilitate searches for new atmospheric constituents (examples of these arc noted below).

#### **Kcy ATMOS** Results

ATMOS has so far been flown on three Spacelab flights. It was first flown on Spacelab 3 in 1985 and has since been flown on the first two flights in the ATLAS series of missions, Results from these three flights will be discussed in turn, highlighting some of the scientific achievements.

### Spacelab 3

The first ATMOS shuttle mission as part of the Spacelab 3 payload was flown on the Space Shuttle "Challenger", launched on April 29th, 1985. On this first flight, a pressure leak in the instrument's reference laser housing limited ATMOS operations to a 24 hour period between April 30' and May 1st, after which the reference laser could not be restarted, During that period data were obtained through 13 complete at mospheric sunset observations centered around 30"N latitude and six complete sunrise observations around 47°S<sup>(2)</sup>. This was a modest operational success compared with later flights, but a huge scientific success nonetheless.

Among the spectral features identified arising from atmospheric constituents were those of a number of gases for which this data set provided the first detection in the middle atmosphere. Included in these were three important reservoir species of the NO.

family ClONO<sub>2</sub><sup>(3)</sup>, N<sub>2</sub>O<sub>5</sub><sup>(4,5)</sup>, and HNO<sub>4</sub><sup>(6)</sup>. Together with measurements of HNO<sub>3</sub>, NO<sub>2</sub>, and NO, the ATMOS SL-3 data provided a complete inventory of the NO<sub>x</sub> family<sup>(7)</sup> including C10NO<sub>2</sub>, the key link between the NO<sub>x</sub> and ClO<sub>x</sub> families, The ATMOS data were also used to measure profiles HCl, the final sink species for atmospheric chlorine, up to heights of 60 km<sup>(8)</sup>. These measurements, together with the other chlorine-containing gases (including all of the most abundant halogen source gases), have provided an estimate of the chlorine loading of the atmosphere for 1985<sup>(9)</sup> - an upper limit to the amount of Cl available for ozone destruction.

In addition to the main species<sup>(10)</sup>, the quality of the spectra returned permitted the retrieval of the profiles of a large number of isotopes of the minor gases<sup>(11)</sup> which promise to provide a sensitive means of testing the importance and the rate of atmospheric processes through the resulting fractionation detected in certain isotopomers. However, it should be emphasized that this first data set as a whole provided a unique ability of to test current photochemical models of the stratosphere to reproduce the observed levels of key gases, as well as simpler assumptions such as the steady-state relationships between various species<sup>(12)</sup>18).

Although not directly associated with the primary objectives of the ATMOS investigation, sufficient high signal-to-noise high resolution infrared solar spectra were obtained during the Spacelab 3 mission to publish the highest quality atlas of the infrared spectrum of the Sun yet produced, A companion volume to this atlas was also published of the middle infrared telluric spectrum at various altitude intervals in the Earth's atmosphere,

# ATLAS-1

Between March 24<sup>th</sup> and April 2nd, 1992, ATMOS flew as part of the ATLAS-1 payload on the Space Shuttle "Atlantis". The payload also included a number of other atmospheric remote sensing experiments and solar measurement instruments providing a

comprehensive set of measurements for the study of global change<sup>(19)</sup>. The shuttle was launched at 13:01 GMT into a 57° inclination orbit, and during the flight, ATMOS was successfully operated through some 94 occultation events covering Latitudes from 30°N to 55°S.

Most noticeable during this flight was the clear effects of the residual stratospheric aerosol created by the Mt. Pinatubo volcano eruption of July, 1991, the largest such event this century in terms of material injected into the stratosphere. The ATMOS spectra showed broad spectral features not present in the earlier SL-3 data that coincide with extinction caused by H<sub>2</sub>SO<sub>4</sub>/H<sub>2</sub>O droplets. It is these droplets, on the order of microns or less in diameter, which provided the means for stimulating unusual photochemical pathways which ATMOS measurements were uniquely able to confirm. The reaction:

$$N_2O_5(g) + H_2O(aerosol) \rightarrow 2HNO_3(g)$$

was suggested to account for the high levels of HNO<sub>3</sub> observed following the eruption. ATMOS were was the only instrument on the ATLAS-1 flight able to provide measurements of both the decrease in N<sub>2</sub>O<sub>5</sub> and concomitant increase in HNO<sub>3</sub><sup>(21)</sup>. How this removal of nitrogen oxides (NO and NO<sub>2</sub> are readily exchanged to N<sub>2</sub>O<sub>5</sub>) has effected the other families of gases, particularly CIO<sub>x</sub>, is a current topic of importance, as it is now clear that 1992 and 1993 were extremely unusual in the low global levels of stratospheric ozone which have been measured.

occurring seven years after the Spacelab 3 mission, this set of new measurements provided a unique opportunity to measure the change in the stratospheric chlorine-loading. While measurements from the ground have been able to monitor the rate of change, these ATMOS measurements were able to determine the absolute change in the concentration of chlorine present at the critical altitudes around the ozone layer. Mixing ratios of 3,44 ppbv and 1.23 ppbv for HC1 and HF above 50 km were measured which, when compared 10 the measured

values obtained on the 1985 flight, correspond to a 37% and 62% increase for HCl and HF. respectively<sup>(22)</sup>. The derived trend in HCl is in good agreement with the model-predicted increase in chlorine loading of 0.13 ppbv year <sup>1</sup>. We attribute the main source of this change to the release of man-made chlorofluorocarbons (CFCs) and hydrochlorofluorocarbons (HCFCs) whose production and usc arc now controlled by international agreement in the Montreal Protocol and its subsequent revisions, London and Copenhagen, Model calculations based on these measured levels predict the growth and subsequent decline of atmospheric chlorine to span several decades before returning to the critical value of below 2 ppbv at which the Antarctic ozone "hole" is believed to have been initiated, Future measurements of HCl and HF can be used as an accurate measure of chlorine loading, and will help monitor what kind of chemicals arc being used to replace the CFCs.

#### ATLAS-2

A night launch of the Space Shuttle "Discovery" on April 8, 1993, gave ATMOS an opportunity on its third flight to obtain measurements at orbital sunrise at high northern latitudes (between 60° to 68°N). Although the flight was delayed beyond the critical period when cold temperatures in the lower stratosphere result in the formation of polar stratospheric clouds and the formation of active chlorine (i.c. Cl<sub>2</sub> and then ClO), the polar vortex was still intact at launch and was positioned over northern Siberia, These fortuitous circumstances provided ATMOS the opportunity to obtain profiles of its inventory of gases inside the residual polar vortex as well as outside,

Preliminary results indicate that many of the signature conditions identified at lower altitudes during aircraft measurement campaigns were still prevalent in the vortex, including a) low values of the source gases N<sub>2</sub>0, CFCs, CH<sub>4</sub> etc. following descent of stratospheric air during the earlier winter period, and b) massive conversion of available inorganic chlorine to ClONO<sub>2</sub>. This data is currently being analyzed and incorporated

into models to test theories of the photochemical formation of and recovery from the stratospheric polar winter conditions.

# Conclusions

The time scale for some of the global changes now occurring as a result of industrial or other anthropogenic activities are clearly measured in decades. Some of these changes arc relatively small, such as global warming, but some, such as the reduction in stratospheric ozone, can readily be discerned against the background of variations in atmospheric state seen between seasons. interannually, or caused by major natural events. To further understand the magnitude of these changes requires measurements on a global scale and over a significant period of time. In the long term, this will probably be met by a concerted set of instruments deployed on free-flying satellites, such as those envisaged for the Earth Observing System (EOS). Currently, these arc scheduled for some time in the future. in the intervening period, and concurrently with the operation of the EOS satellites, shuttle-borne instruments can continue to build on the valuable data base of measurements started almost a decade ago. This will provide continuity, and a proven, calibrated series of observations which cannot be mate.heel by other means,

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